

Suppression of hidden order in URu₂Si₂ under pressure and restoration in magnetic field

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We describe here recent inelastic neutron scattering experiments on the heavy fermion compound URu₂Si₂ realized in order to clarify the nature of the hidden order (HO) phase which occurs below $T_0 = 17.5$ K at ambient pressure. The choice was to measure at a given pressure P where the system will go, by lowering the temperature, successively from paramagnetic (PM) to HO and then to antiferromagnetic phase (AF). Furthermore, in order to verify the selection of the pressure, a macroscopic detection of the phase transitions was also achieved in situ via its thermal expansion response detected by a strain gauge glued on the crystal. Just above $P_x = 0.5$ GPa, where the ground state switches from HO to AF, the $Q_0 = (1, 0, 0)$ excitation disappears while the excitation at the incommensurate wavevector $Q_1 = (1.4, 0, 0)$ remains. Thus, the $Q_0 = (1, 0, 0)$ excitation is intrinsic only in the HO phase. This result is reinforced by studies where now pressure and magnetic field H can be used as tuning variable. Above P_x , the AF phase at low temperature is destroyed by a magnetic field larger than H_{AF} (collapse of the AF $Q_0 = (1, 0, 0)$ Bragg reflection). The field reentrance of the HO phase is demonstrated by the reappearance of its characteristic $Q_0 = (1, 0, 0)$ excitation. The recovery of a PM phase will only be achieved far above H_{AF} at $H_M \approx 35$ T. To determine the P-H-T phase diagram of URu₂Si₂, macroscopic measurements of the thermal expansion were realized with a strain gauge. The reentrant magnetic field increases strongly with pressure. Finally, to investigate the interplay between superconductivity (SC) and spin dynamics, new inelastic neutron scattering experiments are reported down to 0.4 K, far below the superconducting critical temperature $T_{SC} \approx 1.3$ K as measured on our crystal by diamagnetic shielding.

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INTRODUCTION

URu₂Si₂ is a heavy fermion compound with a body centered tetragonal crystal structure in its paramagnetic phase. At ambient pressure, on lowering the temperature below $T_0 = 17.5$ K a transition occurs into a hidden order phase which cannot be a conventional magnetic phase. Over the last decade, the picture was that of small moment antiferromagnetism (as all neutron scattering experiments point out a tiny ordered moment $M_0 \approx 0.02\mu_B$ with the propagation vector $Q_{AF} = (0, 0, 1)$ [1]) associated with an electronic instability corresponding to a loss of electronic carriers at T_0 [2, 3, 4]. The departure from the conventional AF picture is clearly demonstrated by the fact that the entropy loss at T_0 , $\Delta S \approx 0.2R \ln(2)$, is too high to be accounted for exclusively by the formation of localized tiny ordered moments. NMR and μ SR spectroscopy rule out an intrinsic origin of the tiny sublattice magnetization [5, 6]; the small moment seems generated by lattice defaults. This high sensitivity to lattice imperfections is clearly related with the observation that at a rather weak pressure $P_x \approx 0.5$ GPa [7, 8] the ground state switches from HO to AF ground state with a substantial sublattice magnetization $M_0 = 0.4\mu_B$ at $T \rightarrow 0$ with the same Q_{AF} . Here we tune from HO to AF under pressure to clarify the main properties of each phase and in particular noting their inelastic neutron scattering spectra.

At the opposite to the elastic AF neutron scattering

response at ambient pressure, the inelastic neutron scattering signals are very robust [1]: excellent agreement exists between results. Below T_0 , inelastic neutron scattering experiments indicate two sharp and intense excitations, one with a symmetric shape in the energy spectrum at the incommensurate position $Q_1 = (1.4, 0, 0)$ with a gap $E_1 \approx 4.8$ meV and another one with an asymmetric shape at the wavevector $Q_0 = (1, 0, 0)$ with a gap $E_0 \approx 2$ meV. Note that the wavevector $Q_0 = (1, 0, 0)$ is in another Brillouin zone equivalent to the ordering wavevector $Q_{AF} = (0, 0, 1)$. On warming above T_0 , both excitations respond differently. The signal at Q_0 is quasielastic and strongly damped, whereas at Q_1 it remains inelastic but so strongly damped that the spectrum is not fully gapped any more. The T -evolution of the excitation at Q_1 and equivalent positions can explain the specific heat anomaly at T_0 [9]. It is worthwhile to recognize that if no phase transition occurred at T_0 , URu₂Si₂ would be a classical intermediate valence compound; it is the feedback between spin dynamics and band structure reconstruction which leads to the fact that below T_0 sharp excitations at Q_0 and Q_1 emerge [10].

RESULTS AND DISCUSSION

Figure 1 shows the (T, P) phase diagram of URu₂Si₂ as recently determined by resistivity and microcalorimetry experiments [11]. Special attention was given in the de-

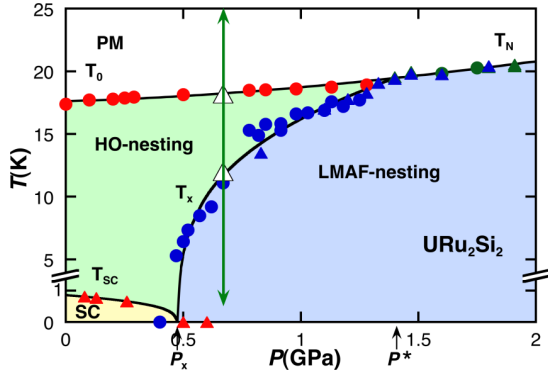


FIG. 1: Pressure-temperature phase diagram of URu_2Si_2 determined by specific heat and resistivity measurements [11]. The green vertical line indicates the pressure where the excitations were measured.

termination of the three lines: $T_0(P)$ of the transition from PM to HO, $T_x(P)$ from HO to AF and $T_N(P)$ from PM to AF. In agreement with thermal expansion and elastic neutron scattering experiments [7, 8, 13], $P_x \approx 0.5$ GPa at $T \rightarrow 0$, while the three lines seem to meet at the critical pressure $P_c \approx 1.3$ GPa with $T_c \approx 18.5$ K. The P -evolution of the two excitations were reported nine years ago [14]; four different pressures were measured on crystals where P_x was reported near 1.5 GPa. Both excitations exist at 0.87 GPa but disappear simultaneously at 1.86 GPa. Taking into account progress in the determination of the (P, T) phase diagram and also our recent ability to measure simultaneously neutron scattering and thermal expansion via a strain gauge in the same pressure cell, we decided to study at a selected pressure P between P_x and P_c (vertical green line in figure 1) the evolution of the inelastic neutron scattering spectrum in the three phases PM, HO and AF for both Q_0 and Q_1 using the high performance of the respective triple axis spectrometers IN12 and IN22 at the Institut Laue Langevin (ILL) [12]. The figure 2 shows the results for the two wavevectors. Clearly, the collective excitation at $Q_0 = (1, 0, 0)$ is characteristic of the HO phase as it disappears below $T_x \approx 13$ K while the signal at $Q_1 = (1.4, 0, 0)$ remains but shifts in energy. Considering the fact that the $Q_0 = (1, 0, 0)$ excitation is characteristic of strong longitudinal fluctuations at the wavevector Q_{AF} and that no drastic difference is detected in the carrier number between both sides of P_x [11], the proposal is that the Fermi surface reconstruction at T_0 and T_N are identical. This statement is supported by our recent study of Shubnikov-de Haas oscillations through P_x [Hassinger *et al.* to be published], where it was observed that the three observed frequencies (α, β, γ) are P invariant. In the AF phase the two uranium sites are inequivalent and the symmetry changes. In excellent agreement with band structure calculations [15, 16, 17], a drop of carrier den-

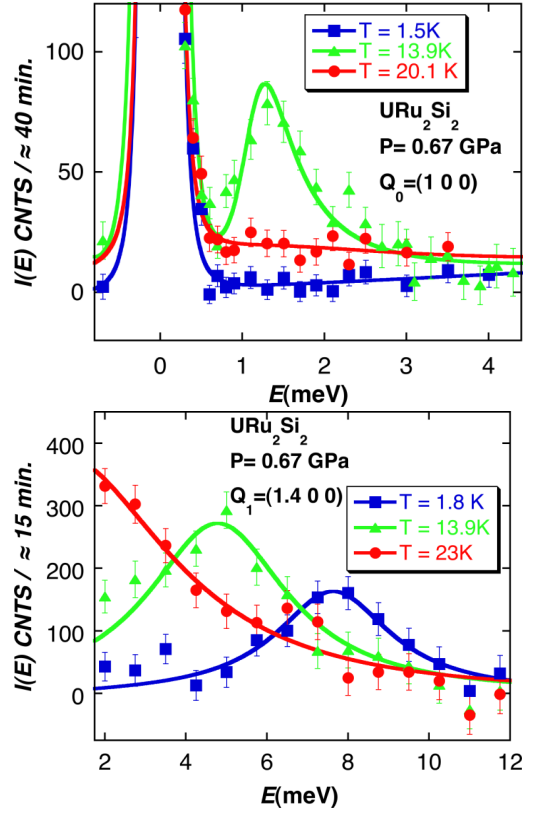


FIG. 2: On the upper panel are shown the energy spectra at the wave vector $Q_0 = (1, 0, 0)$. The excitation present at 13.9 K in the HO state (green triangles) disappears completely at low temperature in the AF phase (blue squares). On the lower panel the excitation at $Q_1 = (1.4, 0, 0)$. The lines are guides to the eye.[12]

sity is associated with this change. In the HO phase, our measurements strongly suggest that Q_{AF} will also be the wavevector of the HO state. Recently it was stressed out from band structure calculations, that the strong longitudinal fluctuations at Q_{AF} in the HO phase preserve a Fermi surface reconstruction similar to the AF state [15].

A recent complementary experiment was to continue the inelastic neutron scattering studies with the addition of a magnetic field [18]. The experiments were performed on the IN14 spectrometer at the ILL and restricted to the field evolution of the Q_0 excitations. The reduction of signal by the combined use of a pressure cell in a cryomagnet device (high magnetic field up to 14.5 T) led us to prepare a large crystal of 6 mm diameter and 11 mm length oriented with the easy c magnetization axis along the magnetic field H ; the selected transmitted medium was a mixture of fluorinert FC 84/FC 87. As before, the selected pressure $P \approx 0.72$ GPa was verified via the detection of the phase transitions PM-HO-AF via the thermal expansion measured by strain gauge. Figure 3 shows the temperature dependence of the magnetic Bragg peak intensity (measurement of the intensity at the

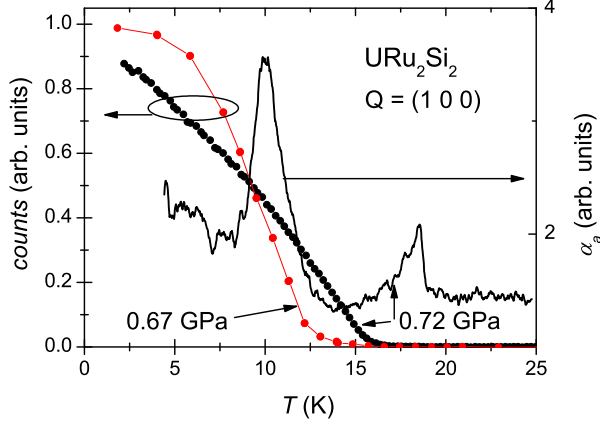


FIG. 3: Scattering intensity at the magnetic Bragg peak position as a function of temperature at a fixed pressure of 0.72 GPa in zero field (black dots, left scale) compared to the same signal in reference [12] (red dots + line) at 0.67 GPa. Additionally the temperature derivative of the strain gauge resistance (black line, right scale) is shown, which is proportional to the thermal expansion coefficient α_c and our indicator for phase transitions. [18]

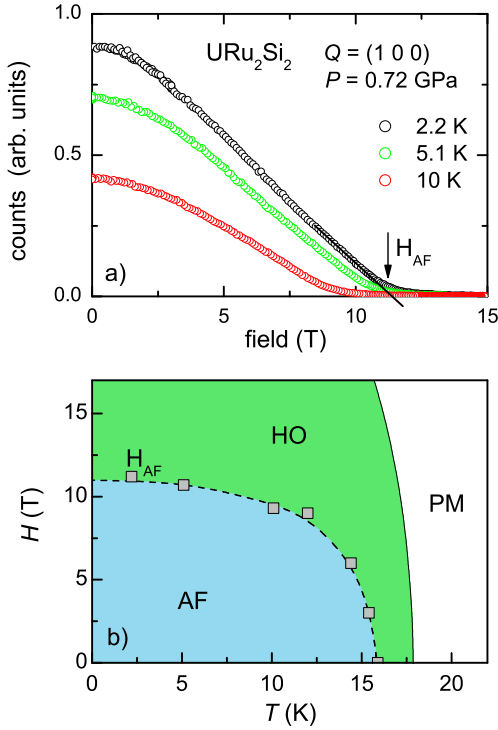


FIG. 4: a) Field dependence of the intensity of the magnetic Bragg peak at $Q_0 = (1, 0, 0)$ at three temperatures at $P = 0.72$ GPa. Above H_{AF} , the AF phase is completely destroyed. b) The phase diagram obtained from the curves on the left panel and from temperature sweeps at constant fields. The transition line between HO and AF follows the behavior measured by thermal expansion at 0.75 GPa interpolated with high field data at 0.8 GPa [12, 18, 19].

$Q_0 = (1, 0, 0)$ position) at zero field at 0.72 GPa (black dots). By comparison, the signal obtained with the previous pressure experiments is drawn as well as the relative variation of the thermal expansion coefficient. It is obvious that pressure inhomogeneities exist in the present set up and that furthermore the deviation from hydrostatics is larger than the previous set up. From the H and T evolution of the Bragg reflection, we can estimate that 10% of "parasitic HO" phase remains even at $T = 0$ and $P = 0.72$ GPa while ideal conditions will lead to the complete disappearance of the HO component as in the previous measurement (see figure 2, left panel). However, following the field evolution of the intensity of the Bragg reflection at different temperatures (figure ??a), a complete disappearance of AF occurs above $H_{AF}(T = 0) = 11$ T. The figure ??b represents the temperature evolution of the field $H_{AF}(T)$ where AF disappears.

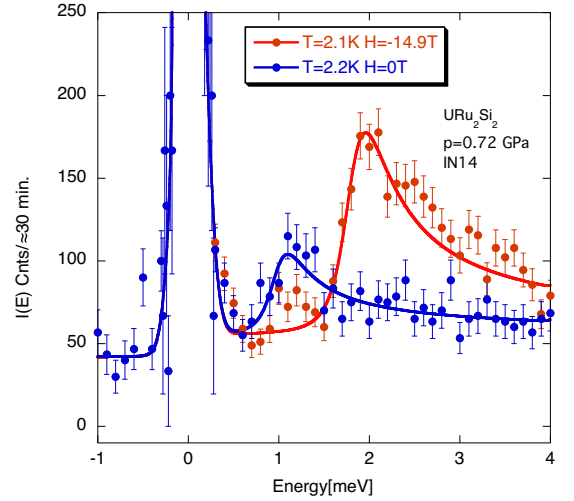


FIG. 5: Energy spectra at the wave vector $Q_0 = (1, 0, 0)$ without magnetic field (blue dots) and in 14.9 T (red dots) at 0.72 GPa. The lines are guides to the eye.[18]

Inelastic neutron scattering experiments lead to the interesting result that a nice excitation reemerges above H_{AF} for $Q_0 = (1, 0, 0)$ (shown in figure 5), the weak contribution detected at $H = 0$ being a consequence of the 10 % parasitic HO survivance. Taking into account our previous P study, our proposal is that the disappearance of AF is associated with the field reentrance of the HO phase. Increasing further the magnetic field, the HO will be destroyed above $H_M \approx 35$ T (the field to suppress the HO at zero pressure is $H_M \approx 35$ T and increases with pressure [19]). In this high field domain, the original higher carrier number of the PM phase will be recovered and different phases occur[20].

Neutron scattering experiments are crucial to point out the nature of the (P, H) induced phases, however it is very arduous to realize a systematic pressure study. This was achieved by detecting the (P, H) evolution of the transition temperatures by thermal expansion via a

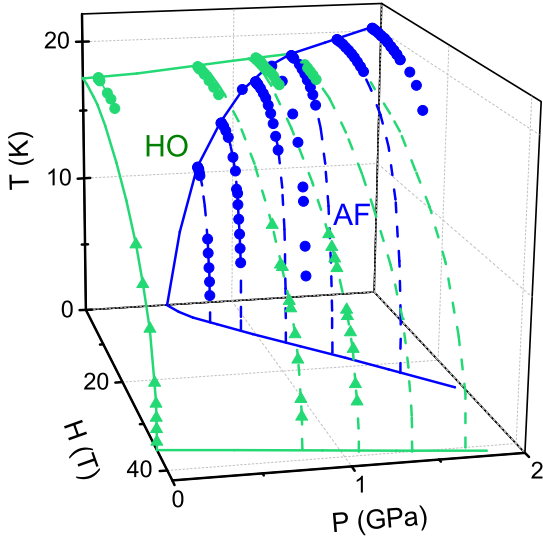


FIG. 6: Three dimensional phase diagram from thermal expansion measurements [18]. The high field data (green triangles) is from reference [19]. The lines are guides to the eye.

strain gauge as represented in figure 6.

At P_x bulk superconductivity collapses as well as the excitation at $Q_0 = (1,0,0)$. Surprisingly, no inelastic neutron scattering experiment at this wavevector has been reported in the SC phase. That led us to conduct an accurate inelastic scattering experiment with a high quality crystal ($T_{SC} \approx 1.2$ K from the measurement of its diamagnetic shielding) down to 0.4 K on IN12 spectrometer at ILL [21]. The temperature dependence of the inelastic spectrum for both Q_0 and Q_1 excitations were recorded. Figure 7 shows the intensity of the inelastic response at Q_0 for $T = 2$ K ($> T_{SC}$) and $T = 0.4$ K ($< T_{SC}$). At 2 K, no quasielastic contribution can be detected at low

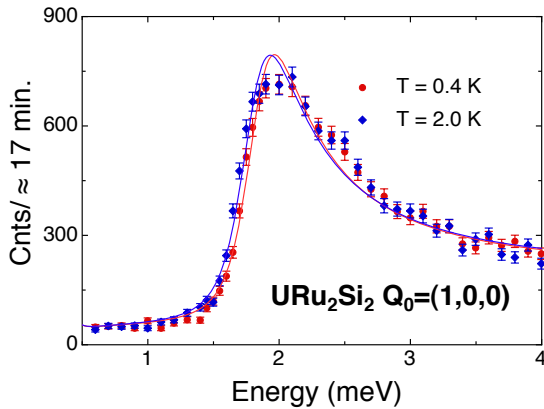


FIG. 7: Energy spectra at the position $Q_0 = (1,0,0)$ above the superconducting transition at 2 K and below it at 0.4 K at ambient pressure. The lines are guides to the eye.

energy; the same lack of quasielastic component was observed for the energy response at Q_1 . At a first glance,

no drastic change occurs below T_{SC} at $T = 0.4$ K for both wavevectors Q_0 and Q_1 : no emergence of a low energy resonance characteristic of the superconducting pairing as observed for the heavy fermion superconductors UPd_2Al_3 , CeCu_2Si_2 and CeCoIn_5 [22, 23, 24, 25]. Looking more closely, the excitation at Q_0 shifts by $40 \mu\text{eV}$, while the excitation at Q_1 remains unchanged. The shift in energy of the excitation at Q_0 through T_{SC} as well as the collapse of the inelastic response above P_x may suggest that the origin of the SC pairing can be magnetic excitations as proposed for UPd_2Al_3 .

The absence of detectable, low energy quasielastic ($T > T_{SC}$) or resonant ($T < T_{SC}$) contributions at Q_0 by comparison to UPd_2Al_3 , CeCu_2Si_2 and CeCoIn_5 can be explained partly by the weakness of its Sommerfeld coefficient γ ($\gamma = 70 \text{ mJmol}^{-1}\text{K}^{-2}$ in URu_2Si_2 , $140 \text{ mJmol}^{-1}\text{K}^{-2}$ in UPd_2Al_3 , about $1000 \text{ mJmol}^{-1}\text{K}^{-2}$ in CeCu_2Si_2 and CeCoIn_5 [26]). Indeed a weak γ corresponds to a large relaxation rate for the normal state fluctuations ($T > T_{SC}$) and therefore the excitation spectrum is spread out in energy space. It is also possible that this magnetic response in URu_2Si_2 is not strongly structured in momentum space as well contrarily to the three previous cases where the quasielastic response is concentrated along the antiferromagnetic hot spot. The case of URu_2Si_2 appears rather similar to that of the skutterudite system $\text{PrOs}_4\text{Sb}_{12}$ where despite a rather large γ -term ($\approx 300 \text{ mJmol}^{-1}\text{K}^{-2}$) no quasielastic contribution has been detected around any wavevector [27, 28]. For the case of $\text{PrOs}_4\text{Sb}_{12}$, it was recently stressed that the mass enhancement may originate from the aspherical Coulomb scattering of conduction electrons from the very low energy singlet-triplet crystalline electric field excitation [29]. This mechanism is related to the dominant quadrupolar degrees of freedom at play in this compound. Multipolar degrees of freedom may be involved in the formation of complex hidden order phase in URu_2Si_2 .

On entering under pressure above P_x in the AF phase, it was observed from resistivity measurements that the gap energy jumps to a higher value at P_x [18]. According to P studies realized on 1-1-5 heavy fermion systems such as CeRhIn_5 on the duality between AF and SC, this jump can explain the collapse of SC in the AF phase [30]. Thus the proof that the $Q_0 = (1,0,0)$ excitation may be the source of the SC pairing deserves more theoretical treatment.

CONCLUSION

New neutron scattering experiments show clearly that the excitation at $Q_0 = (1,0,0)$ is characteristic of the hidden order state. It disappears under pressure at P_x where the ground state switches from HO to AF and is restored above H_{AF} where the groundstate switches from AF to HO. There are strong indications from spin dy-

namics, but also from transport measurements that the wavevector of the HO phase is $Q_{AF} = (0, 0, 1)$. It is also demonstrated that the Q_0 excitation is directly coupled with superconductivity (P and T variation). Experimentally, URu₂Si₂ is a beautiful case where the main effect at the first order transition at P_x occurs on the sublattice magnetization, on the inelastic neutron scattering response and on the thermal expansion, while other probes like the bulk magnetization, the resistivity, the Shubnikov-de Haas oscillations have only minor changes [31]. For neutron scattering, the next targets are to determine the relation between the sharp excitations at Q_0 and Q_1 and the Fermi surface reconstruction. Theoretical work on the absence of quasielastic response, on the source of SC pairing and on the (P - H) instability of HO and AF phases is strongly needed.

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